Nonproliferation Nuclear Forensics

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Abstract   Nuclear terrorism is one of the most serious security threats facing the world today. Many countries, including the United States, have incorporated nuclear forensic analysis as an important part of their strategy to prevent nuclear terrorism and nuclear smuggling. Nonproliferation nuclear forensics involves the laboratory analysis of seized illicit nuclear materials and the interpretation of data to identify the origin and intended use of the material. Forensic signatures have been developed to improve the confidence with which nuclear forensic scientists can draw conclusions. These signatures are validated and new signatures developed through research and development programs and in round-robin exercises among nuclear forensic laboratories worldwide. Nuclear forensics involves an iterative approach based on the application of a wide array of state of the art analytical techniques. Studies of interdicted materials conducted in cooperation with government or law enforcement agencies provide the ultimate test for analytical protocols developed under controlled laboratory conditions. We illustrate the current state of nuclear forensics using two case studies of nuclear material.

Introduction.   Beginning with the breakup of the Soviet Union in the early 1990s, unprecedented amounts of illicitly obtained radiological and nuclear materials began to be seized at border crossings and international points of entry. The first instances of this new criminal activity, "nuclear smuggling", were reported in 1991 in Italy and Switzerland and in subsequent years numerous incidents involving illicit trafficking of radioactive or nuclear material occurred in a number of central European countries. Between 1993 and 2011, the International Atomic Energy Agency (IAEA) recorded more than 2150 incidents of illicit trafficking of radioactive material [IAEA, 2012] More than 400 of these incidents involve *bona fide* nuclear material, primarily depleted, natural or low-enriched uranium. Of special concern, moreover, are the 16 or so events involving highly-enriched uranium or plutonium (Table 1). This overt evidence of significant amounts of nuclear material outside lawful control has created international concern over the importance of maintaining global nuclear order and underscores President Obama's statement in

Prague in 2009, "In a strange turn of history, the threat of global nuclear war has gone down, but the risk of nuclear attack has gone up."

Out of the need not only to identify and characterize illicit nuclear materials but also to learn more about both the original and intended use of the material, its origin and the putative trafficking route, a new scientific discipline was developed, "nuclear forensics." In the U.S., the nuclear forensics effort was jump started by taking advantage of several decades of experience developed through the nuclear weapons program, supplemented with expertise from geochemistry, material science and conventional forensics.

Nuclear forensics is the technical means by which intercepted radioactive or nuclear material (and any associated non-nuclear material) are characterized to determine, e.g., the chemical and isotopic composition, physical state and age; these data are then interpreted to evaluate provenance, production history and trafficking route. The goal of these analyses is to identify forensic indicators in the interdicted nuclear and radiological samples or the surrounding environment, e.g., container, transport vehicle or packaging. These indicators arise from known relationships between material characteristics and process history. Nuclear forensics requires a combination of technical data, relevant databases, and specialized skills and knowledge to generate, analyze, and interpret the data. When combined with law enforcement and intelligence data, nuclear forensics can suggest or exclude potential origins and thereby contribute to attribution of the material to its source or production facility.

A primary objective of nuclear forensics is to identify the sources of stolen or illicitly trafficked nuclear materials and thereby prevent or make more difficult terrorist acts that would use material from these same sources [Mayer et al., 2007; Moody et al., 2005]. The perception of effective nuclear forensics is likely to deter some of the individuals who would need to be involved in any act of nuclear terrorism and provides incentives to states to better guard their materials and facilities. The science of nuclear forensics provides the tools and knowledge by which nuclear materials are characterized and interpreted.

The terrorist attacks on New York City and Washington, DC, on September 11, 2001, greatly increased the visibility of nuclear forensics, as policy makers worldwide became increasingly concerned about the possibility of terrorist groups, such as Al Qaeda, obtaining a nuclear weapon or using a radiological dispersal device (RDD or so-called "dirty bomb"). Over the past several years a consensus has developed among international leaders that the threat of nuclear terrorism poses a real and present danger to both national and international security. President Obama, the leaders of 46 other nations, the heads of the International Atomic Energy Agency

and the United Nations, and numerous experts have called nuclear terrorism one of the most serious threats to global security and stability. The Communiqué of the 2012 Seoul National Security Summit recognizes that nuclear forensics can be an effective tool in determining the origin of detected nuclear and other radioactive materials and in providing evidence for the prosecution of acts of illicit trafficking and malicious uses. Furthermore, the Communiqué encourages States to work with one another, as well as with the IAEA, to develop and enhance nuclear forensics capabilities and underscores the importance of international cooperation both in technology and human resource development to advance nuclear forensics [Communiqué, 2012].

A basic challenge facing nuclear forensics is the application of modern material analysis techniques, knowledge of commercial and military nuclear fuel cycles, and scientific principles to analyze unknown nuclear materials or devices and provide information of value to decision-makers. This problem is complex enough before considering the wide range of potential materials that may be encountered and the many different types of information that potentially may be required. As in classical forensics, nuclear forensics relies on the fact that certain measurable parameters in a sample are characteristic for a given material. Using these characteristic parameters, also known as "signatures", nuclear forensic analysis seeks to draw conclusions on the origin and intended use of the intercepted material.

The technical response to specific nuclear incidents requires a graded, iterative approach. "Categorization" addresses the threat posed by specific interdicted material by identifying the risk to first responders, law enforcement personnel, and the public, and then determining whether or not there is an indication of criminal activity or threat to national security. "Characterization" provides a more thorough analysis of the material to determine the nature of the radioactive and associated, non-nuclear evidence. "Nuclear forensic interpretation" seeks to draw validated technical conclusions from the analytical results, correlating the characteristics of the material with material production history. While nuclear forensic interpretation is the end product for the nuclear forensic laboratory, it provides input into the nuclear attribution process. Complete nuclear forensic analysis includes characterization of all materials, traditional forensic analysis, and nuclear forensic interpretation. This approach, predicated on the model action plan developed by the Nuclear Forensics International Technical Working Group (ITGW), is described in much greater detail in the IAEA publication, "Nuclear Forensics Support," Nuclear Security Series Number 2 [Smith et al., 2008; IAEA, 2006].

Nuclear forensic interpretation is a deductive process (e.g., Fig. 1), much like the scientific method itself. Initially, nuclear forensic scientists develop an hypothesis, or set of hypotheses, based upon the initial analytical results. In most cases, these

initial results will be consistent with multiple hypotheses, which may, in turn, suggest additional signatures. The team then develops additional measurements to verify the presence or absence of the signatures. If analyses show that the signature is absent, this hypothesis must be rejected or adjusted to fit the new results. If, instead, the analyses confirm the signature, then either the investigation has come to a unique technical interpretation, the desired result, or additional tests to exclude other remaining hypotheses must be developed. In the ideal case, only a single hypothesis or interpretation will eventually prove consistent with all results, although this is seldom true in practice.

Signatures. Nuclear forensic scientists use the term "signatures" to describe material characteristics that may be used to link samples to people, places, and processes, much as a written signature can be used to link a document to a particular individual. "Signatures" describe any characteristic or group of characteristics that can be used to help distinguish materials from one another or identify the processes history of a material. Signatures are essentially combinations of variables used to make comparisons. Some signatures, such as those associated with U or Pu isotopic analyses, may provide only general clues that serve to place the material in a broad category, e.g., Depleted Uranium (DU) or Highly Enriched Uranium (HEU), or, perhaps, narrow the field of potential countries of origin. Other signatures, such as characteristic dimensions or markings, are generally applicable to only a restricted class of materials, e.g., reactor fuel elements or sealed sources, but may provide valuable clues identifying a specific facility or date of manufacture. In some cases, data generated in a nuclear forensic investigation may provide useful information only when combined with other, complementary results. Signatures for nuclear materials are intimately connected to the nuclear fuel cycle. Each step in the fuel cycle (Fig. 2) both creates new signatures and erases or modifies some existing signatures. In general, materials at each stage of the fuel cycle will have their own characteristic signatures. An on-going challenge for nuclear forensics is to validate signatures for each step in the fuel cycle and understand the processes that control a signature's persistence.

Almost without exception, a single signature is insufficient to answer all of the relevant questions. Independent signatures that reach the same conclusion increase confidence in the technical interpretation, while results that provide different or conflicting conclusions decrease the level of confidence. Nuclear forensic investigations are most successful when independent signatures representing a variety of material characteristics can be linked and point toward a unique conclusion. Figure 3 illustrates this process schematically by depicting the universe of potential nuclear material sources and processes. Each individual signature defines a subset of know materials from which an intercepted sample may have

originated. In the idea case, the use of multiple signatures leads to a unique point of intersection of multiple subsets corresponding to a unique identification of the source and/or process. Overall, we should aim to gather as many material characteristics as possible since this gives us the richest possible set of signatures on which to make comparisons.

Signatures generally fall into two broad categories: comparative signatures and predictive signatures. Comparative signatures involve the comparison of the measured properties (e.g., grain size, color, chemical and isotopic composition) of an unknown sample (or "questioned sample" in law enforcement parlance) to a similar set of properties for one or more reference samples. The essential question to be addressed is whether or not the characteristics of an unknown sample are the same as, or at least are similar to, one or more of the reference samples. The use of comparative signatures to identify an interdicted sample may involve either a point-to-point comparison or a point-to-population comparison. Point-to-point comparisons are relatively rare and rely on the inter-comparison of two or more closely matched samples, e.g., HEU interdicted in Bulgaria in 1999 and in Paris in 2001 [Adamson et al., 2001; Baude, 2008]. Point-to-population comparisons look for similarities between the characteristics of an unknown sample and those of a population of potentially similar materials and are more broadly applicable. Point-to-population comparisons usually require access to databases containing information on hundreds or thousands of samples, or to nuclear forensic sample archives, which may contain tens or hundreds of physical samples. The value of the comparative approach then depends strongly on the relevance and coverage of the database and/or sample archive (see, e.g., Dolgov et al., 1999; Robel et al., 2009).

Predictive signatures, in contrast, come into play when representative data for a suite of appropriate reference materials are unavailable. Predictive signatures typically derive from underlying scientific principles, such as isotopic and chemical fractionation, in the case of uranium ore and ore concentrate, neutron capture activation and fission, in the case of nuclear reactor modeling, or radioactive decay in the case of age dating. Predictive signatures seek to calculate material characteristics useful for attribution based on a detailed understanding of the physical or chemical mechanisms responsible for producing the signatures. The advantage of the predictive approach is that the processes (and possibly locations) of unanalyzed nuclear materials can be inferred from their measured characteristics, something of critical importance for types of materials that are not readily available, e.g., materials from historical processes or tightly-held materials from foreign countries. The disadvantage of the predictive approach is that significant effort must be expended to develop and validate the capability and to understand accurately the processes affecting signatures.

New predictive signatures can also be developed through advances in the understanding of the processes affecting chemical and isotope distributions at the molecular, atomic and nuclear scale. The $^{234}U/^{238}U$ ratio, for example, exhibits substantial variability in water, soil, sediment and uranium ore samples of different geographical origin [Gascoyne, 1992] ^{234}U is preferentially leached compared with ^{238}U from solids due to radiation damage of the crystal lattice from alpha decay of ^{238}U, oxidation of insoluble tetravalent ^{234}U to soluble hexavalent ^{234}U, and alpha recoil of ^{234}Th (and its daughter ^{234}U) into fluid phases. Ores leached by groundwater over long periods of time exhibit significant depletions in ^{234}U, whereas ores formed through deposition of those water leachates exhibit complementary enrichment in ^{234}U; the full range in ^{234}U concentration is nearly 20% [Brennecka et al., 2010]. Modern mass spectrometry provides results of sufficient precision and accuracy to allow small variations in the $^{238}U/^{235}U$ ratio, once thought to be invariant in nature, can to be measured. The depositional environment of an ore body appears to strongly influence the $^{238}U/^{235}U$ ratio with low-temperature ores having systematically higher ratios than deposits formed at higher temperatures [Tumey et al., 2009]. In addition, naturally occurring variations in ^{236}U content can also be exploited as a nuclear forensic signature. Generally considered to be an anthropogenic isotope, ^{236}U is produced at very low levels in U-ore bodies through neutron capture on ^{235}U; the abundance of ^{236}U is strongly influenced by the age of the ore-body and the volume of water in contact with ore [Tumey et al., 2009; Wilcken et al., 2008]. All these features of the isotopic distribution of natural uranium are potentially useful (predictive) signatures for attribution of U-ore and ore concentrate.

Nuclear Forensic Analysis. Nuclear forensic analysis does not lend itself to a simple "cook book" approach, universally applicable to all types of nuclear and radiological material. Instead, nuclear forensics involves an iterative approach, in which the results from one analysis are used to guide subsequent analyses. The international nuclear forensics community has defined 3 levels of analysis – categorization, characterization, and full nuclear forensic analysis – each of which serves a specific purpose in an investigation. In all cases, though, sampling and analysis must be performed with due regard for preservation of evidence and chain-of-custody requirements. Many of the analytical tools used in these analyses are destructive and consume some amount of sample during analysis. Proper selection and sequencing of analyses is, therefore, critical.

The goal of **categorization** is to identify the bulk constituents of a sample to assess the threat posed by the material and confirm whether the interdicted material is contraband and is likely to form the basis for continued investigation. Categorization should occur on-site, at the point of interdiction and utilize non-

destructive analytical techniques such as field-portable gamma-ray spectrometry, and hand-held x-ray fluorescence. These nondestructive analyses can quickly distinguish between naturally occurring radioactive material, special nuclear material, radioactively contaminated material, or a commercial radioactive source.

The goal of **characterization** is to determine the nature of the radioactive evidence. Characterization provides full elemental analysis of the interdicted material, including major, minor and trace constituents. For major constituents of the radioactive material, characterization should also include determination of isotopic and phase (i.e., molecular) properties. Characterization also includes measurement of physical properties, including accurate measurement of critical dimensions of solid samples, determination of particle size and morphology for powder samples, and high magnification imaging of the material by optical and scanning electron microscopy.

The goal of **full nuclear forensic analysis** is to analyze all radioactive and traditional forensic evidence to gather information to address questions of material origin, method of production, loss of legitimate control, transit route from point of diversion to interdiction, and likelihood that additional material is available. Full nuclear forensic analysis also includes detailed nuclear forensic interpretation and often includes comparison of measured signatures against information contained in nuclear forensics databases or sample archives or against predictive signatures generated by, e.g., reactor modeling, to assist in the identification of the method of manufacture and most plausible source of the material.

Nuclear forensics employs a wide array of analytical tools to detect signatures in radioactive material. The international nuclear forensics community has achieved a general consensus on the proper sequencing of analytical techniques to provide the most valuable information as early as possible in a nuclear forensic investigation. This consensus was achieved through discussion and consultation at regular meetings of the ITWG, as well as from experience developed through three round robin analyses by nuclear forensic laboratories. The collection of time or environmentally sensitive samples should occur within the first 24 hours after interdiction. Non-destructive analysis should be conducted before destructive analyses whenever possible. Table 2 shows the generally accepted sequence of analysis, broken down into techniques that should be performed within 24 hours, 1 week, or 2 months after interdiction. Table 3 provides an overview of many analytical techniques commonly used in nuclear forensic investigations; additional information may be found in [Moody et al., 2005].

Radiometric Techniques. Radiometric techniques measure the radiation radioactive nuclides emit when they decay to a daughter nuclide. There are three types of

radiation commonly encountered in nuclear forensics - alpha, beta and gamma radiation, each with its own properties and methods of detection. Most heavy nuclides (e.g., U and Pu) decay by emitting an alpha particle. Gamma radiation is also often emitted after the alpha decay to bring the daughter nuclide from an excited state to the ground state. Each nuclide emits characteristic gamma rays with energies specific to an individual radioisotope. While useful for characterizing the performance of chemical separations in the laboratory, β spectrometry is rarely employed as a quantitative technique. Most β -emitting radionuclides also emit γ rays characteristic of the decaying nuclides; however, a few radionuclides (including long-lived fission products such as ^{99}Tc and ^{147}Pm and other nuclides of potential interest such as ^{14}C and ^{3}H) β-decay without accompanying photon emission.

Gamma spectroscopy has a dual role in nuclear forensics. It is the first technique that is used when interdicted nuclear material is investigated. Since gamma rays are only slightly attenuated by packaging material (unless shielding like lead is used), initial measurements in the field (e.g., at border crossing stations) carried out with simple, portable gamma spectrometers provide rapid and accurate categorization of the material to distinguish, for example between naturally occurring radioactive material, radioactive source, medical isotopes or anthropogenic nuclear material. In laboratories, more sophisticated high-resolution gamma spectrometers (HRGS) are used. Their energy resolution is much better than in the portable instruments, allowing gamma rays with energies very close to each other to be resolved. HRGS provides an initial determination of the isotopic composition of uranium and/or plutonium, as well as detection and quantification of trace fission and activation products. It should, however, be noted that some nuclides like ^{242}Pu or ^{236}U cannot be detected by gamma spectroscopy. In these cases, mass spectrometry offers a useful alternative.

Alpha spectroscopy is used to quantify the abundance of alpha-emitting radionuclides, particularly those with relatively short half-lives. Alpha particles are stopped for example by a paper sheet, because of their strong interaction with matter. Consequently, an alpha measurement through packaging material or shielding is impossible. Unlike gamma spectroscopy, alpha spectrometry is a destructive technique requiring rather laborious sample preparation. Source preparation is crucial for achieving good energy resolution in alpha spectroscopy and target elements are usually separated and purified before being deposited onto a flat surface. Quantification is achieved by spiking the samples with known amounts of an isotopic spike or tracer. Alpha spectrometry is especially suited for quantifying ^{232}U and ^{238}Pu due to their short half-lives and, in the case of ^{238}Pu, the potential interference from ^{238}U in mass spectrometry. Alpha spectrometry is also used to quantify ^{241}Am (daughter of ^{241}Pu), whose concentration can then be used to

calculate the date of the last plutonium purification performed on a sample and ^{230}Th (daughter of ^{234}U), to determine a last purification date for uranium materials.

X-ray diffraction (XRD) is the standard method for identifying the chemical structure of crystalline materials. X-ray beams impinging on regularly ordered lattices undergo constructive and destructive interference depending on the spacing of the lattice, the wavelength of the X-rays, and the angle of incidence of the x-ray beam. By rotating a sample relative to a fixed X-ray source, variations in interference lead to characteristic diffraction patterns. These diffraction patterns can be compared to reference spectra to identify specific crystalline phase. XRD is not applicable to amorphous (non-crystalline) materials.

X-ray fluorescence (XRF) provides non-destructive quantification of chemical concentrations in both solid samples and solutions for elements from Mg to Pu. An incident beam of high energy x-rays excites characteristic secondary x-rays whose intensities are quantified using wavelength or energy sensitive detectors. The detection limits for XRF are generally in the range of tens of μg /g, although actinide matrices generate many x-rays that interfere with lower-energy x-rays of lighter elements. Wavelength dispersive analysis (WDS) provides higher energy resolution than energy dispersive analysis (EDS), and can be used to resolve some of these interferences. XRF is often used as a screening tool in nuclear forensic analyses to guide additional analyses using mass spectrometry.

<u>Mass spectrometry.</u> Mass spectrometric techniques make use of the mass differences between nuclides. In mass spectrometry the atoms contained in a sample are converted to ions and then separated according to their mass to charge ratio and the intensities of the mass-separated ion beams measured. Mass spectrometry is used to determine both the elemental and isotopic compositions of nuclear materials, providing extremely high precision and accuracy, as well as the capability to analyze both radioactive and stable isotopes. Mass spectrometry can provide quantification of elemental concentrations either by using an isotopic spike (isotopic dilution mass spectrometry) or through calibration against standard solutions in the case of solution-based techniques like inductively coupled plasma mass spectrometry (ICP-MS). Nuclear forensic analysis utilizes a variety of different types of mass spectrometers, which differ primarily in the way ions are generated. One important exception is accelerator mass spectrometry, which accelerates ions to MeV energies rather that the keV energies used in most mass spectrometers.

In Thermal Ionization Mass Spectrometry (TIMS), samples consisting of small (~fg–μg) quantities of chemically separated and purified analytes dissolved in a small volume (typically 1–10 ml) are deposited on a refractory metal filament (e.g., high-purity W or Re) and evaporated to dryness. The filament is then heated to

temperatures of 1,000–2,500°C in the ion source by resistive heating. If the ionization potential of the analyte is low compared to the work function of the filament, a fraction (typically <1%) of the analyte atoms will be ionized and emitted from the filament surface. Multi-collector TIMS instruments, with detectors able to measure multiple isotopes simultaneously, are capable of measuring differences in isotope abundance ratios as small as a few parts in 10^6. TIMS is the preferred technique for measuring Sr and Nd isotopes and U and Pb isotopes with the highest possible precision and accuracy.

A disadvantage of TIMS is the laborious sample preparation. As in the case of alpha spectrometry, samples need to be dissolved and chemically purified to avoid mass interferences and achieve high sensitivity. In many laboratories TIMS has been replaced by inductively coupled, plasma-source mass spectrometers (ICP-MS). In ICP-MS, a sample is dissolved in an acid solution, which is nebulized in a spray chamber and aspirated into an Ar plasma. The analyte dissociates into atomic constituents and ionizes in the high temperature plasma (5,000–8,000 K) with very high efficiency (>90% for elements with a first ionization potential of <8 eV). The salient features of ICP-MS are multi-element capability, high sample throughput, good sensitivity and large dynamic range. Multi-collector ICP-MS instruments (MC-ICP-MS) provide high precision and accuracy measurements of Pb, U and Pu isotopic compositions. In addition to measuring isotopic compositions, ICP-MS is a powerful and widely applied method for quantifying trace element abundances. The minimum detection limits for MC-ICPMS are generally <1 pg/g and can reach the fg/g range for favorable elements.

Laser-ablation, inductively coupled-plasma mass spectrometry (LA-ICPMS) uses a laser-ablation cell and optical microscope to supplant the spray chamber/nebulizer of a standard ICP-MS instrument. Material is ablated from a sample using a laser (often a pulsed Nd-YAG tuned to 266 or 213 nm) and transported in a stream of Ar gas to the plasma torch for ionization and subsequent mass analysis as per solution ICP-MS. Only minimal sample preparation is required. While laser spot sizes can be reduced to several micrometers, sensitivity is degraded as a result, and typical spatial resolution is ~10–100 μm. Matrix matched standards are necessary for accurate trace element and isotope analyses in LA-ICPMS. Depending on the quality of standards, LA-ICPMS accuracy is typically 0.2-5% with limits-of-detection in the ng/g range. The combination of laser ablation and MC-ICP-MS can produce data with much higher precision and accuracy [Arevalo et al., 2010].

Secondary ion mass spectrometry (SIMS) is a microanalytical technique, applicable to samples ranging in size from centimeters to submicron particles and providing both elemental and isotopic information. A finely focused primary ion beam (e.g., O+ or Cs+) hits the sample and sputters atoms from the surface. SIMS uses a finely

focused primary ion beam, e.g., O^+_2, O^-, Cs^+, or Ga^+, to sputter the sample surface, producing secondary ions that are then analyzed by a mass spectrometer. SIMS is capable of acquiring microscopic images of isotopic and elemental distributions with spatial resolution as high as 50 nm. SIMS can be used to measure the concentration of any element, from H to Pu, over a dynamic range of more than nine orders of magnitude. SIMS is applied in nuclear forensics when only small amounts of sample (e.g., particles) are available or when the sample is inhomogeneous and individual constituents are to be investigated. The sputtering process is very matrix-dependent and accurate quantitation requires matrix-matched standards. The precision and accuracy of isotope-ratio measurements is typically 0.05-0.5%. SIMS is the technique of choice to determine isotope ratios and trace element abundances in particulate samples. Using a sharply focused primary ion beam, SIMS can analyze particles in the μm-size range, weighing <1 pg, with a precision and accuracy of better than 0.5%. In many nuclear forensic applications, a few U- or Pu-bearing particles may be immersed in a sea of environmental detritus containing little in the way of forensic information. SIMS can be used in a particle-search mode to locate and analyze these rare, but highly valued, particles. As with laser ablation ICP-MS, the adoption of large geometry multi-collector mass spectrometers has significantly improved SIMS capabilities, particularly for determination of low-abundance isotopes like ^{236}U [Ranebo et al., 2009].

Gas chromatography-mass spectrometry (GC-MS) is a technique for detecting and measuring trace organic constituents in a bulk sample. In GC-MS, the components of a mixture are separated in a gas chromatograph and identified in a mass spectrometer. The primary component of a GC is a narrow-bore tube maintained inside an oven. In the simplest arrangement, the analyte mixture is flash-vaporized in a heated injection port. The various components are swept through the column by a carrier gas and separated on the column based upon relative absorption affinities. In an ideal case, components elute from the column separated in time and can be introduced into the mass spectrometer as a time series. The mass spectrometer detects and quantitates the concentration of each component as it elutes from the column. GC-MS analyses provide very high specificity, allowing extremely complex mixtures to be accurately separated and individual species to be identified accurately. Limits of detection for scanning GC-MS are on the order of ng of material, corresponding to sensitivities of ~1 part in 10^{13} for simple samples and 1 part in 10^{11} for complex mixtures.

Imaging Techniques. The role of microscopy is to provide a magnified image of a sample, allowing the observation of features beyond the resolution of the unaided human eye (roughly 50-100 μm). The ability to identify and characterize diverse

suites of samples rapidly and without compromising the integrity of the sample is an essential starting point of most forensic investigations. A variety of microscopy techniques are applied in nuclear forensic science, using photons, electrons, and x-rays to probe the physical, chemical, and structural make-up of samples at spatial scales ranging from nanometers to centimeters.

Optical microscopy dates back > 300 years and remains one of the most basic and fundamental characterization techniques in nuclear forensics. The optical microscope is often the first instrument used to examine a sample in detail, and allows the forensic scientist to answer the simple, yet vital, question, "what does the sample look like?" before proceeding with more extensive, and often destructive, analyses. Ranging from simple hand lenses to sophisticated compound microscopes, optical microscopy reveals details of color, surface morphology and texture, shape and size, tool marks, wear patterns, surficial coatings, corrosion, and mineralogy [Grant et al., 1998; Sokova et al., 2008]. The stereomicroscope produces three-dimensional images at relatively low magnification (~2-80×) and is very useful for dissecting or aliquoting samples for additional analyses. The polarizing microscope passes light through a set of polarizing filters to gain additional information about the nature of the sample from optical properties such as crystallinity, anisotropy, pleochroism and birefringence; polarizing microscopes can readily magnify an image to 1000×. The limit of resolution is set by the wavelength of light used to illuminate the sample; the theoretical resolution limit of conventional microscopes is 200 nm, but values closer to 1 μm are more commonly achieved.

In Scanning Electron Microscopy (SEM), a finely focused electron beam is rastered over a sample and the interaction of the incident electron beam with the sample produces a variety of signals: backscattered electrons, secondary electrons, Auger electrons, x-rays, and photons. By measuring the intensity of one or more of these types of particles as a function of raster position, an image of the sample is constructed and displayed. Each type of emitted particle conveys different information about the sample, and, by choosing the appropriate detection mode, either topographic or compositional contrast is revealed in the image. Secondary electrons arise from inelastic collisions between incident electrons and atomic electrons within the outer few nm of the surface and carry information about sample topology (e.g., Fig, 4). Backscattered electrons, in contrast, have energies comparable to the incident electron beam, carry information about the mean atomic number and can be used to construct maps of the distribution of phases with disparate chemical composition. With thermionic, W filament sources, image resolution is limited to ~10 nm, with a corresponding maximum magnification of 100,000. With field-emission electron sources, the resolution exceeds 1 nm with a

corresponding maximum magnification of 1,000,000.

Transmission electron microscopy (TEM) lies at the opposite end of the spectrum from optical microscopy – difficult to use and requiring elaborate sample preparation. Its unique capabilities for ultra-high spatial resolution and for revealing microstructural information, however, make the TEM an important tool in many nuclear forensic investigations. In TEM, a high-energy electron beam is transmitted through a very thin sample (< 300-nm thickness). In the imaging mode TEM produces a magnified image of the sample providing information on thickness, crystallinity, crystal orientation, defects and deformations. Understanding how contrast is generated is key to distinguishing among these competing effects and presents a significant challenge in image interpretation. The diffraction mode provides an electron diffraction pattern, analogous to an x-ray diffraction pattern. Electron diffraction patterns can be indexed by the same procedures used in x-ray diffraction and used to identify phases on an extremely fine spatial scale. Just as in SEM, characteristic x-rays are generated by the interaction of the electron beam with the sample. X-ray analysis can be combined with TEM imaging and diffraction to provide comprehensive information on a specimen's internal microstructure, with nm spatial resolution. TEM is capable of an extremely wide range of magnification (from ∼ 50× to several million×) and is able to image extremely fine structural detail, but at the expense of severe restrictions on sample thickness.

The characteristic x-rays generated by interactions between energetic electrons and the sample in SEM or TEM carry information on chemical composition and provide an important method to determine elemental concentrations for most solid samples, including micrometer size particles. Characteristic x-rays are emitted as a result of electron transitions between inner shells of the constituent atoms in a specimen and emitted as "lines" with precisely defined energies, corresponding to different elements. The characteristic x-rays can be analyzed by either of two methods. An energy-dispersive x-ray spectrometer (EDS) uses the photoelectric absorption of x-rays in a semiconductor detector, usually Si(Li), to measure the energy and intensity of incident x-rays simultaneously. EDS systems provide an easy-to-use method of measuring x-ray spectra over a broad energy range and can detect elements from B to U. Detection limits are typically ∼0.1% for silicate and oxide materials. A wavelength-dispersive spectrometer (WDS) operates on the principle of Bragg diffraction; x-rays are dispersed according to wavelength, rather than energy. WDS provides much higher energy resolution (50-100×) and sensitivity (∼10×) compared to EDS. WDS will detect elements from Be to Pu, with detection limits of 0.01%. X-ray microanalysis is particularly valuable in nuclear forensic investigations for the speed with which x-ray intensities can be accurately quantified to yield elemental concentrations in interdicted samples.

Infrared Spectroscopy (IR) is useful for the identification of organic compounds. Through the use of an infrared microscope, IR can be performed on samples as small as 10 μm and is an important micro-analytical technique in nuclear forensics. Molecular bonds vibrate at characteristic frequencies and if a particular molecular vibration results in a change in the bond's dipole moment, the molecule can absorb infrared radiation corresponding to that characteristic frequency. In IR, a sample is irradiated with a broad band of infrared frequencies and the intensity of the reflected or transmitted radiation is measured as a function of frequency. Absorption at specific frequencies is characteristic of specific bonds and the IR spectrum identifies the various bonds and functional groups within the molecule. Extensive libraries of IR spectra help identify unknown compounds but unambiguous identification usually requires the use of an additional analytical technique, such as mass spectrometry or NMR.

Chronology. Radionuclides linked to one another by radioactive decay have relative concentrations that can be calculated by the simple laws of radioactive in-growth or, in more complicated cases, by the Bateman equations. The measurement of the relative concentrations of parent and daughter isotopes provides a direct measure of the time since the daughter radionuclides were removed from the respective parent isotopes. In nuclear forensic investigations, the interval between the time a sample was purified and the time it was subsequently analyzed is defined as the "age" of the material.

The presence of both U and Pu provides the opportunity to measure the age of a sample through as many as a dozen different chronometers. If the ages given by different chronometers "agree" with each other (concordant ages), then we have high confidence the assumptions for accurate age dating are satisfied and the model ages reflect the time since purification. If the chronometers do not agree with each other (discordant ages), caution must be exercised in the way model ages are interpreted, as they may fail to indicate accurately the time since purification. Table 4 lists the quantities of heavy-element daughter nuclides present in a 1-gram sample of weapon-grade Pu after an in-growth period of one year. If the sample was completely purified during the last chemical separation, all of the chronometers should yield the same age. However, while the ^{232}U – ^{236}Pu, ^{234}U – ^{238}Pu, ^{235}U – ^{239}Pu and ^{236}U – ^{240}Pu chronometers all yield the same age in most U.S. weapons-grade Pu metal samples, ^{241}Am – ^{241}Pu often gives a significantly larger value. This discordance indicates that when U was last removed from the Pu, some Am was left behind. As a result, there will be more ^{241}Am in the sample than can be explained by in-growth, resulting in an apparent age that is too large.

Case Studies

The ultimate test for protocols developed in the laboratory in a controlled environment is posed by their application to real world samples, obtained under uncontrolled conditions and whose properties often contain unexpected features. Case studies are normally conducted in cooperation with government or law enforcement agencies with responsibility for sample collection. The agency responsible for collecting the sample works with the nuclear forensic scientists to develop a Statement of Work (SOW) specifying the material properties to be measured. In most cases the SOW follows the nuclear forensics Model Action Plan described in IAEA *Nuclear Security Series #2* [IAEA, 2006]. The SOW also lays out the time lines for analysis and reporting of final results.

Counterweight – A Nuclear Smuggling Hoax. A dense, dark gray ~9-kg metal sample was involved in a sale of illicit nuclear materials in Hong Kong in 1988. The sample had been offered for sale as "nuclear weapon useable material" by a Southeast Asian military official and was subsequently rediscovered in a U.S. consulate nearly 10 years later. LLNL was contacted with a request for forensic characterization (a photograph may be found in Grant et al., 1998).

HRGS analysis revealed that the main radioactive component of the specimen was U, considerably depleted in ^{235}U. Bulk analysis of the sample yielded a density of (17 ± 0.3) g/cm^3, somewhat less than the theoretical density of U-metal. The reduced density of the part suggested that voids could be present, or that it was composed of two or more inhomogeneous phases.

After consultation with the collecting agency, the sample was characterized using radiochemical analysis, electron microprobe, SIMS, ICP-MS and XRF. The results showed that the material was depleted U containing ~ 0.3 wt.% ^{235}U and was an alloy of 90% U with 10% Mo. The sample was coated with electroplated Ni ranging in thickness between 85-150 μm. The crenulated outer margin implied that the piece had been cast and not been machined prior to Ni plating. Radio-chronometry based on ^{234}U – ^{230}Th determined the date of last chemical purification as 1961 (± 3 years).

Once the nuclear forensic information was collected, LLNL carried out a complementary investigation using conventional forensics and determined the part had been made by the National Lead Company of Albany, NY and then transferred to Nuclear Metals, Inc. The interdicted specimen was most plausibly a piece of an aircraft counterweight assembly, most plausibly from a U.S. military aircraft.

In the nuclear smuggling world, this sample was one of the earliest contraband items in what ultimately became known as the "Southeast Asian Uranium" scam. This hoax is a pervasive swindle, first reported in 1991, and especially prevalent in Thailand, Vietnam, and Cambodia. Transactions of irregularly shaped metal parts,

alleged to be ^{235}U, with asking prices of ~$10,000 per item, are not uncommon. Similar material has also been used for barter as substitute currency in drug trafficking operations.

<u>High Enriched Uranium Interdicted in Bulgaria.</u> Just after midnight on 29 May 1999, a Turkish citizen, Urskan Hanifi, was stopped at a border crossing in Ruse, Bulgaria on his way into Romania. Although claiming to be returning from an extended trip to Turkey, the Bulgarian border guard became suspicious because the car's interior was very tidy and appeared to contain no luggage. A search of the car turned up a certificate for the purchase of "99.99% uranium 235" written in Cyrillic and a lead container labeled "uranium 235" concealed inside an air compressor in the trunk of the car. Inside the container was a glass ampoule filled with several grams of fine black powder that Bulgarian scientists confirmed to be highly enriched uranium (Fig. 5). Hanifi then tried to bribe the customs officials, who refused his money and instead arrested him. According to press reports, Mr. Hanifi, told police he had purchased the uranium in Moldova and had been trying to sell it in Turkey; having failed, he was attempting to return to Moldova.

Roughly one year after the uranium-filled vial was seized in Bulgaria, the U.S. Dept. of State arranged for it to be sent to Lawrence Livermore National Laboratory with the hope that detailed analyses could offer clues to the material's origin. Over the next 9 or so months, a team of nuclear forensic scientists from LLNL and several other Dept. of Energy laboratories performed an exhaustive study of the HEU and the associated packaging materials, revealing a wealth of information that ultimately led investigators to the source of the HEU [Adamson et al., 2001].

Following an initial evaluation by HRGS, revealing that the HEU contained ~72% ^{235}U, 1% ^{234}U and no significant Pu, the sample and packaging materials were characterized using optical microscopy, SEM and TEM, both with energy dispersive x-ray analysis, XRD, radiochemistry followed by α- and γ-spectrometry and mass spectrometry, optical emission spectrometry, ion-, gas- and gel- permeation chromatography, GC-MS, IR spectrometry, x-ray photo-electron spectroscopy, and XRF.

The HEU is a very fine-grained powder composed predominantly of U_3O_8. The powder formed loosely compacted clumps ranging to 100 μm in size. Individual particles are irregularly shaped and distinctive morphologies are absent at the resolution provided by the SEM. TEM performed on an aliquot of the sample revealed two distinctive classes of particles. Equant to slightly ovoid grains dominate the population, comprising ~ 90% of the total, with rod-shaped and plate-shaped grains making up the remainder (Fig. 6). A size-frequency analysis of grains showed a wide distribution of sizes, spanning the interval from 30 to 550 nm, with a

mean diameter of only ~160 nm. The abundance of very small grains with diameters of <300 nm, provided an important clue to the manufacturing process used to make the HEU, as such small sizes are difficult to generate by mechanical grinding and milling.

The concentrations of 72 elements, ranging from Li to Th, were measured using a variety of analytical techniques. Individual elements vary widely in concentration, from <2 ng/g to ~200 μg/g. The total impurity inventory, 500 to 800 μg/g, is high compared to other HEU samples, with 4 elements (Cl, S, Fe, and Br) accounting for ~60% of the total inventory. The enrichment of the volatile, electronegative elements, S, Cl, and Br, is most readily interpreted as a signature of chemical reprocessing. Overall, the trace element abundances are much higher than expected for laboratory-scale reprocessing and suggest the HEU is an aliquot of batch reprocessing.

The concentrations of 35 radionuclides, spanning 15 orders of magnitude in concentration, were determined by α- and γ-spectrometry following radiochemical separation. The major constituents are the six U isotopes – ^{238}U, ^{236}U, ^{235}U, ^{234}U, ^{233}U, and ^{232}U – plus ^{230}Th (produced by decay of ^{234}U); ^{241}Am, five Pu isotopes – ^{242}Pu, ^{241}Pu, ^{240}Pu, ^{239}Pu, and ^{238}Pu – ^{237}Np and the fission products – ^{125}Sn, ^{134}Cs, and ^{137}Cs – were also detected. The presence of three fission-products provides incontrovertible evidence the sample is reprocessed U, irradiated in a nuclear reactor.

The isotopic composition of U was determined by three different techniques – SIMS, TIMS, and MC-ICP-MS – and the Pu isotopic composition was determined by TIMS. SIMS provided a rapid (within 24 hours of sample aliquoting), reasonably accurate, initial analysis of the major U isotopes. TIMS and MC-ICP-MS provided data on all six U isotopes, including the low-abundant isotopes ^{232}U and ^{233}U, with much higher accuracy than SIMS or HRGS. The analyses by all of the instrumental methods yielded a consistent U-isotopic composition for the HEU powder, notable especially for the extremely high ^{236}U content. These data are summarized in Table 5. The U isotope abundances identify the material as HEU containing ~73% ^{235}U, i.e., weapon-usable material. The U isotope abundances suggest an initial enrichment of ~90% and the high concentration of ^{236}U indicates a prolonged irradiation history. The isotopic composition of Pu is consistent with weapon-useable material but the concentration (~2 ng/g) is much too low for the HEU to be a significant source of weaponizable Pu.

The age of the sample was determined using nine radio-chronometers, based on the decay of U and Pu. The mean age of the HEU was 6.5 y, relative to the date the radiochemical separation was carried out at LLNL, 17 April 2000, indicating the

HEU was reprocessed on 30 October 1993, with an uncertainty of <1 month. The agreement in age for the nine radio-chronometers indicates that the trace level of Pu in the sample was introduced during reprocessing and is not a recently added contaminant. The ability to determine sample age with high accuracy is significant from a Safeguards perspective. In principle, if the HEU had been diverted from a facility subject to International Atomic Energy Agency oversight, and if reprocessing records were complete, the identity of the sample could be determined on the basis of the accurate age determination alone.

Collateral Evidence. The Pb container was examined via optical and scanning electron microscopy. Marks on the outer surface provided clear evidence of coarse filing by hand for shaping and smoothing; marks indicative of the use of machine tools are absent. The overall appearance of the container, especially the irregular form, suggests the container was cast in a crude sand mold and shaped by hand. This supposition was later confirmed by metallurgical examination.

A small fragment was cut from the container, polished, and etched to reveal the microstructure. The structure, consisting of Pb dendrites surrounded by a two-phase eutectic region, is characteristic of common, cast-Pb metal. SEM/EDS showed ~5 wt.% Sb had been added to the Pb to produce an alloy with greater malleability. The SEM also revealed remnants of alumino-silicate, similar to kyanite (Al_2SiO_5), trapped in the Pb. Kyanite, a naturally occurring mineral, is commonly used for high-temperature insulation and may have been used as a mold wash, liner, or release agent in the casting process.

The yellow wax filling the interior of the Pb container was analyzed by Fourier-transform infrared spectrometry to identify molecular compounds. Based on FTIR spectra, the wax was identified as a paraffin derivative with composition inconsistent with many commercial waxes but was strikingly similar to the paraffin-based wax, Parowax®. The coloring agent was identified using methylene chlorine to extract the paraffin from the inorganic component and then XRF to examine the residue. XRF identified the inorganic residue as barium chromate, $BaCrO_4$, a substance once commonly used as yellow pigment in paints, glass, and ceramic over-glazes, as an oxidizer in pyrotechnics, and as an oxidizer in heat powders and igniters. Barium chromate is rarely used today in the U.S. or most western countries because of environmental and health concerns but widespread use persists in Brazil, China, India, and many eastern European countries.

The two paper samples retrieved from the Pb container were characterized using forensic microscopy to determine the composition of the wood fibers making up the paper. The quality of the paper is similar to commercial office paper. Fibers from the inner paper-liner separating the ampoule from the paraffin wax consisted of

61% bleached softwood and 39% bleached hardwood, while fibers from the label removed from the cap on the shield consisted of 38% bleached softwood, 23% semi-bleached softwood and 39% bleached hardwood. Both the softwood and the hardwood fibers were produced with the Kraft pulping process. The fibers in these papers are not found in North America, Western Europe, or Scandinavia and the papers were most plausibly produced in Eastern Europe.

Attribution. The primary goal of attribution assessment is to identify the original source of the material, the intended, or original, use, and the responsible individual or organization. Attribution typically proceeds in stages, focusing first on unambiguous signatures (e.g., U isotopic composition), then proceeding to more subtle signatures (e.g., trace elements and physical properties), and finally considering collateral signatures found, e.g., in packaging. The dominant signature of the HEU is the U-isotopic composition. The U isotope abundances, especially the unusually high concentration of ^{236}U, clearly indicate the sample is HEU irradiated and then reprocessed reactor fuel. The HEU had an initial ^{235}U abundance of ~90%, immediately excluding material manufactured in the United States, since most U.S. HEU has a ^{235}U content of ~93%. The 90% enrichment is consistent with HEU produced in the former Soviet Union (FSU).

Other characteristics of the fuel and packaging also point to an origin in the FSU. The grain size is characteristic of material prepared for specialized use, e.g., powder metallurgy. The extremely fine grain-size of the powder is unlike that found in U.S. facilities, where coarser sizes are used to minimize the health hazard created by respiration of fine dust. The HEU has the characteristics of feedstock for fabrication of fuel pellets and blending with other batches of uranium oxide at U fuel-conversion facilities in the FSU. Multiple samples, similar to the one discussed here, are commonly taken from batches of uranium oxide product for analysis and archive.

Determining the type of reactor in which the HEU was irradiated is a much more involved process, using knowledge of reactor designs and operating conditions and computer modeling of fuel burn-up. Calculations were performed with the ORIGEN2 code to determine the initial isotope abundances and the reactor neutron spectrum most consistent with the observed uranium isotope abundances. These calculations indicate that a very thermal energy spectrum and a burn-up exposure of ~350,000 MWD/MT best match the measured uranium isotope abundances. The most likely source is a light water reactor, possibly a PWR, test facility or research reactor for naval propulsion systems. The low abundance of ^{241}Pu suggests the fuel was stored for 10-20 y after discharge before reprocessing.

The data for the non-nuclear samples reinforces the assertion that the sample originated in the former Soviet Union. The ampoule has been identified by visitors to FSU nuclear facilities as strongly resembling the glass containers used to preserve aliquots of production runs for archival material. The barium chromate giving the wax its distinctive yellow color is banned in the U.S. and most Western countries but is still widespread in Brazil, China, India and many of the Newly Independent States. The paper products are derived from mixtures of hardwood and softwood trees not found in the U.S. or Western Europe, but common in Eastern Europe. Finally, the Pb isotope composition of the container is inconsistent with Pb mined in the U.S. but compatible with lead from Asia or Eastern Europe.

The preponderance of the evidence thus points to an origin in the FSU. Efforts to refine this attribution analysis are continuing.

Recent interdictions [Sokova and Potter, 2008; Global Security Newswire, 2010; 2011] suggest that attempts to smuggle weapon-useable nuclear materials across international borders have continued. Illicit trafficking in nuclear materials remains an important area of concern for the International Atomic Energy Agency, Europol, and national law enforcement agencies, and has gained increased attention in the context of recent Nuclear Security Summits.

Conclusions

Nuclear forensics is an emerging science, driven primarily by national security objectives, including those of both law enforcement and national intelligence. Nuclear forensics is one input into nuclear attribution, in which responsibility is assigned, along with other sources of information, such as law enforcement and intelligence. Nuclear forensics is used to generate technical conclusions by applying validated signatures to analytical results from the interdicted material. These validated signatures include both comparative signatures, in which the interdicted material is compared to the results from material of known origins, and predictive signatures, in which conclusions are generated without reference to other samples. Valid analytical results, in turn, depend on appropriately validated analytical methods, proper analytical sampling, and a quality control/assurance program.

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Table 1: Selected Interdictions of Nuclear Material

Year	Location	Type	Enrichment or ^{239}Pu content	Mass
1992	Augsburg, Germany	LEU	2.5%	1.1 kg
1992	Podolsk, Russia	HEU	90%	1.5 kg
1993	Vilnius, Lithuania	HEU	50%	100 g
1993	Andreeva Guba, Russia	HEU	36%	1.8 kg
1993	Murmansk, Russia	HEU	20%	4.5 kg
1994	St. Petersburg, Russia	HEU	90%	3.05 kg
1994	Tengen, Germany	Pu	99.7%	6 g
1994	Landshut, Germany	HEU	87.8%	0.8 g
1994	Munich, Germany	Pu	87%	363 g
		LEU	1.6%	120 g
1994	Prague, Czech Republic	HEU	87.8%	2.7 kg
1995	Prague, Czech Republic	HEU	87.8%	0.415 g
1995	Prague, Ceske Budejovice	HEU	87.8%	17 g
1995	Moscow, Russia	HEU	20%	1.7 kg
1999	Ruse, Bulgaria	HEU	72%	4 g
2001	Paris, France	HEU	72%	0.5 g
2003	Ignalina, Lithuania	LEU	2.0%	60 g
2003	Georgia/Armenia Border, Georgia	HEU	~90%	170 g
2003	Rotterdam, Netherlands	NU	0.72%	3 kg
2006	Tbilisi, Georgia	HEU	~90%	80 g
2007	Pribenik-Lacacseke Border, Slovakia	NU	0.72%	426.5 g
2010	Tbilisi, Georgia	HEU	>70%	18 g

Table 2. Time line of analyses in nuclear forensic analyses

Techniques/Methods	24-Hour	One Week	Two Months
Radiological	Estimated total activity Dose rate (α, β, γ, n) Surface Contamination		
Physical Characterization	Visual Inspection Radiography Photography Weight Dimensions Optical Microscopy Density	SEM/EDS XRD	TEM (EDS)
Traditional Forensic Analysis	Fingerprints, Fibers		
Isotope Analysis	γ-spectroscopy α-spectroscopy	Mass spectrometry (SIMS, TIMS, ICP-MS)	Radiochemical Separations
Elemental/Chemical		ICP-MS XRF Assay (titration, IDMS)	GC/MS

Table 3 Examples of Analytical Tools for Nuclear Forensics

Measurement goal	Technique	Type of information	Typical detection limit	Spatial resolution
Survey	HRGS	Isotopic	ng-μg	
Elemental and Isotopic Bulk Analysis	Chemical Assay	Elemental	mg	
	Radiochemistry/Radiometric Methods	Isotopic, Elemental	fg-pg	
	TIMS	Isotopic, Elemental	pg-ng	
	ICP-MS	Isotopic Elemental	pg-ng	
	XRF	Elemental	10 μg/g	
	XRD	Molecular	~1 at%	
	GC/MS	Molecular	μg/g	
Imaging	Visual Inspection	Macroscopic		0.1 mm
	Optical Microscopy	Microscopic Structure		1 μm
	SEM			1 nm
	TEM			0.1 nm
Microanalysis	SIMS	Elemental Isotopic	0.1 ng/g-10 μg/g	0.1 – 1 μm
	SEM/EDS or WDS	Elemental	0.1-1 wt%	1 μm
	FTIR	Molecular	0.1-1 wt%	10 μm
	Raman	Molecular	~1 wt%	1 μm

mg = milligram = 10^{-3} gram
μg = microgram = 10^{-6} gram
ng = nanogram = 10^{-9} gram
pg = pictogram = 10^{-12} gram
fg = femtogram = 10^{-15} gram
at% = atom percent
wt% = weight percent
ppm = parts per million by weight
ppb = parts per billion by weight
μm = micrometer = 10^{-6} meter
HRGS = High-Resolution Gamma Spectrometry
TIMS = Thermal Ionization Mass Spectrometry
ICP-MS = Inductively Coupled Plasma mass Spectrometry
XRF = X-ray Fluorescence Analysis
XRD = X-ray Diffraction Analysis
GC/MS = Gas Chromatography/Mass Spectrometry
SEM = Scanning Electron Microscopy
TEM = Transmission Electron Microscopy
SIMS = Secondary Ion Mass Spectrometry
SEM/EDS = Scanning Electron Microanalysis with Energy Dispersive Sensor
SEM/WDS = Scanning Electron Microanalysis with Wavelength Dispersive Sensor
FTIR = Fourier Transform InfraRed Spectroscopy

Table 4. Abundances of heavy-element daughter nuclides in a 1 g sample of Pu-metal containing 6% ^{240}Pu, 0.91% ^{241}Pu, and 0.023% ^{242}Pu, after 1 year of in-growth.

Nuclide	Half-life (Myr)	Mass (ng)	Activity (dpm)
^{230}Th	0.075	1.3×10^{-3}	0.06
^{231}Pa	0.033	1.3×10^{-5}	0.0013
^{233}U	0.16	5.6×10^{-5}	0.0012
^{234}U	0.25	915	12700
^{235}U	704	26300	126
^{236}U	23.4	6250	897
^{238}U	4470	0.42	3.2×10^{-4}
^{237}Np	2.14	355	555
^{241}Am	4.32×10^{-4}	427000	$3.3e^9$

Table 5. Uranium isotope abundances in HEU interdicted in Bulgaria

Isotope	Abundance (atom %)[1]
^{232}U	$(1.06 \pm 0.06) \times 10^{-6}$
^{233}U	$(3.0 \pm 0.18) \times 10^{-5}$
^{234}U	1.175 ± 0.003
^{235}U	72.657 ± 0.012
^{236}U	12.133 ± 0.004
^{238}U	14.045 ± 0.011

1. Uncertainties are 2 standard deviations.

Figure Captions

1. Flow chart of the Nuclear Forensics analysis and interpretation process.

2. Representation of the nuclear fuel cycle, the progression of nuclear material through a series of stages beginning with the mining of ore; conversion to ore concentrate, UF6, and, finally, U-oxide fuel; irradiation in a nuclear reactor; and reprocessing or disposition of spent nuclear fuel. The inserts show representative images for different stages in the fuel cycle.

3. Diagram showing how individual forensic signatures define the subset of materials from which an interdicted sample may have originated. Applying multiple signatures collectively increases confidence in the assessment of identification of possible origins.

4. SEM photomicrograph of plutonium oxide. The variety of morphologies suggests the sample is a mixture of materials produced under different conditions.

5. Photomicrograph of the HEU interdicted in Bulgaria in 1999. The top image shows the Pb container with its distinctive yellow wax lining, while the bottom image shows the HEU powder inside the glass ampoule. *(Reproduced from Adamson, 2001).*

6. TEM photomicrograph of the HEU seized in Bulgaria revealing two distinctive shapes of grains – oval to equant grains making up ~90% of the sample (left-hand image) and much rarer, elongated, rod- or plate-shaped grains (right-hand image). Both types of grains are U_3O_8. The scale bars represent 300 nm in the left-hand image and 100 nm in the right-hand image. *(Reproduced from Adamson, 2001).*
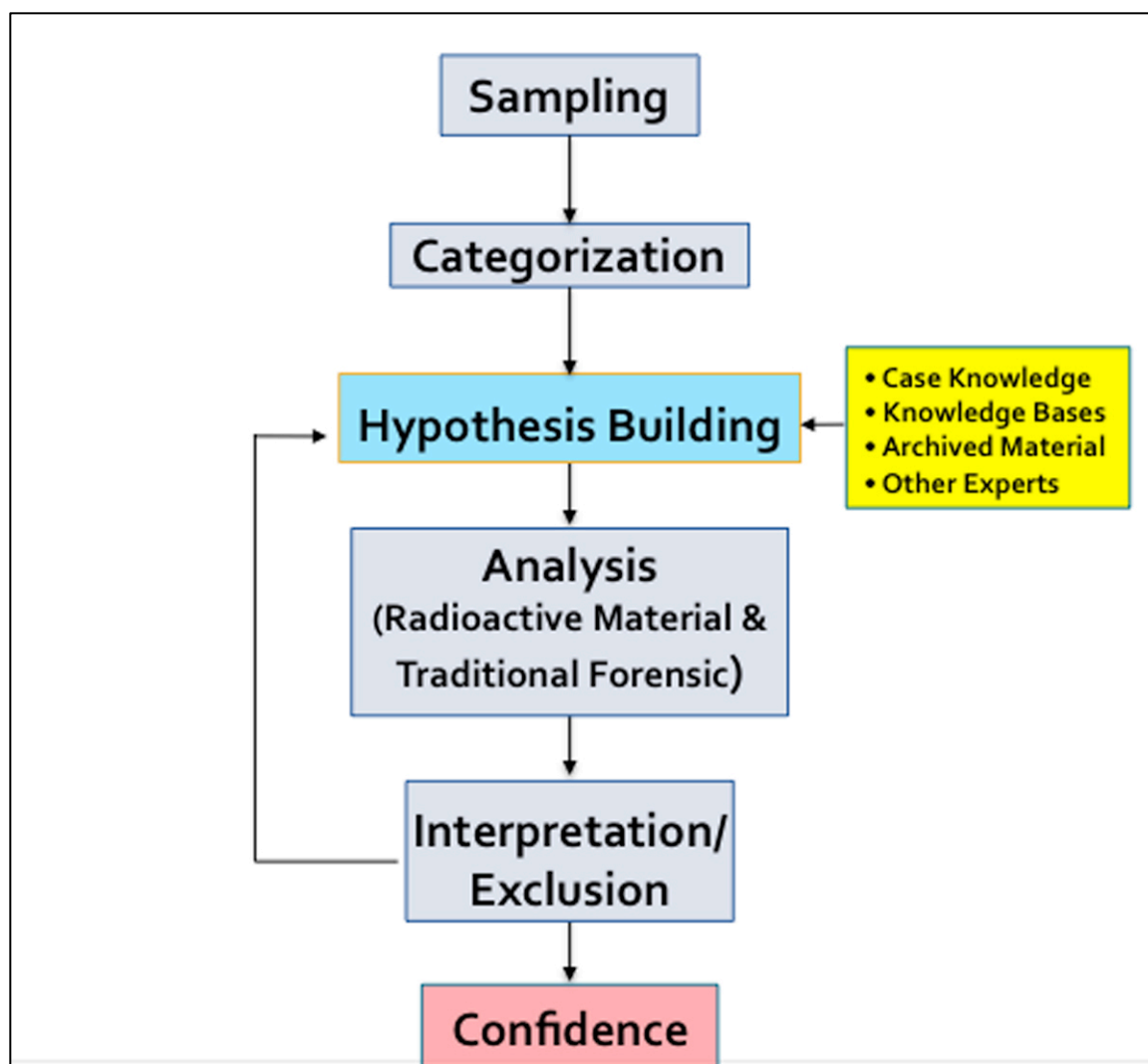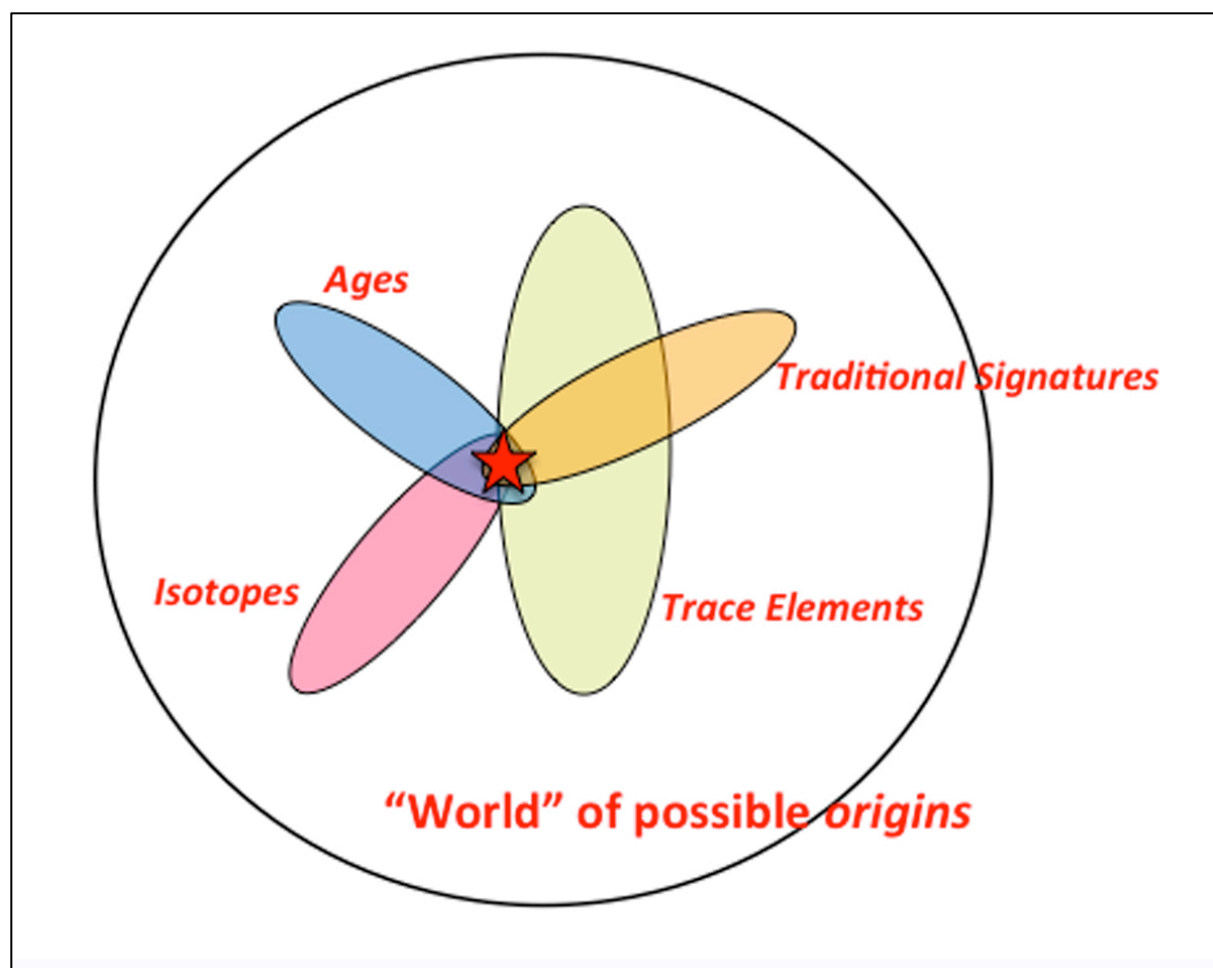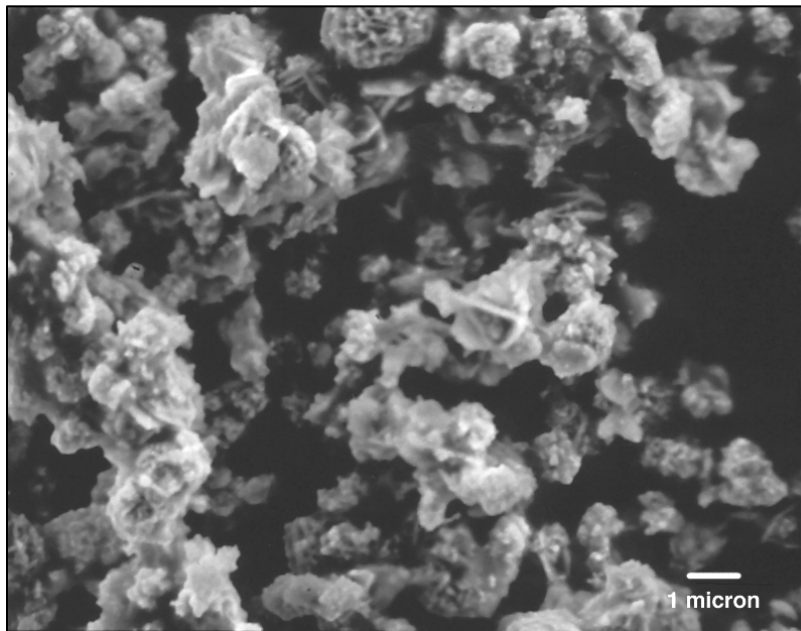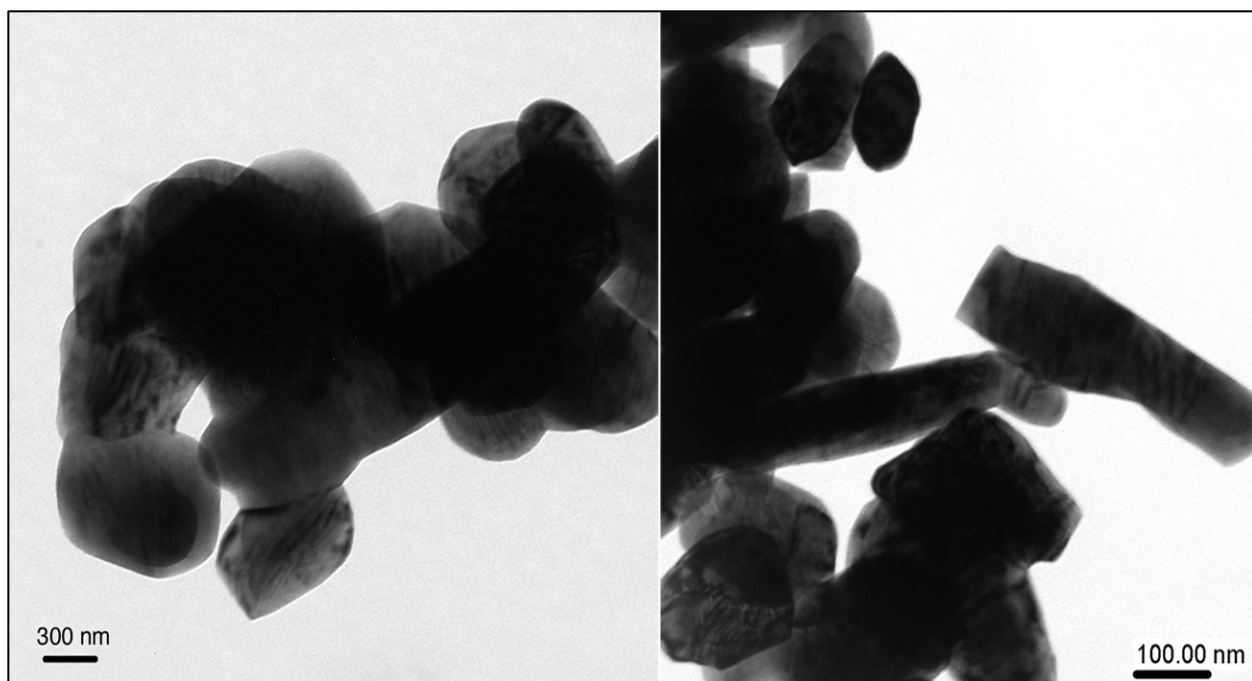
Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6